

PATENT

IN THE U.S. PATENT AND TRADEMARK OFFICE

In re application of

Atsushi FUKAYA et al.

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Application No. 10/594,405

Group 1795

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Examiner Gregg Cantelmo

NONAQUEOUS ELECTROLYTE COMPOSITION AND NONAQUEOUS ELECTROLYTE  
SECONDARY BATTERY USING THE SAME

DECLARATION UNDER 37 CFR 1.132

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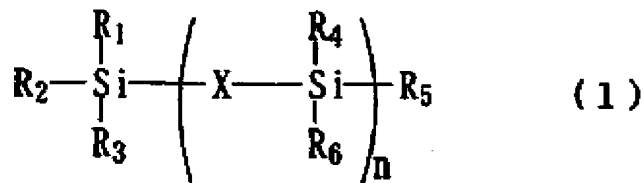
Sir:

I, Takayuki TAKI, am one of the inventors of the above-identified U.S. patent application and declare as follows:

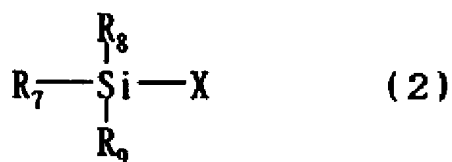
I am familiar with the above-identified application and have read the Official Action dated September 17, 2009, in which the Examiner rejected claim 11 of the present application as being unpatentable under 35 U.S.C. § 103(a) as obvious in view of the combined disclosures of JP 2004-039510 ("Awano") and any one of JP 2003-163032 ("Go"), U.S. Patent Application Publication No. 2003/0113634 ("Oh") and CA 2334054 ("Gan").

The present invention relates to a nonaqueous electrolyte composition which comprises: a mixed organic solvent that includes 20 to 35% by volume ethylene carbonate ("EC"), 35 to

45% by volume ethyl methyl carbonate ("EMC"), 15 to 35% by volume dimethyl carbonate ("DMC") and 3 to 15% by volume either diethyl carbonate ("DEC") or propylene carbonate ("PC"); and a silicon compound represented by general formula (1)



or general formula (2)



Each of Go, Oh and Gan discloses a nonaqueous electrolyte composition comprising a mixed organic solvent, but fails to disclose a silicon compound in accordance with either general formula (1) or general formula (2).

4. U.S. Patent Application Publication No. 2004/0007688 ("Awano") is an English language counterpart to JP 2004-039510. Awano discloses, in Examples 1 and 2 thereof, nonaqueous electrolyte compositions comprising: an organic solvent that includes EC, EMC and DMC; and a silicon compound that is encompassed by general formula (2). Accordingly, of the four references relied upon in rejecting claim 11 of the present

application, Awano should be considered the closest prior art relative to that claim.

The compositions according to the present invention yield superior cycle characteristics, especially low temperature cycle characteristics, in a lithium secondary battery which would have been unexpected by a person of ordinary skill in the art, as is demonstrated by the following comparative test data comparing use of compositions in accordance with the present invention with those disclosed in Awano.

As evidence of the superior and unexpected properties of the present invention, compositions in accordance with the disclosure of Awano were prepared and tested in a lithium secondary battery and compared with the test results obtained using compositions prepared in accordance with the present invention.

Comparative Examples 3-1, 3-2, 3-3 and 3-4

For Comparative Example 3-1, a nonaqueous electrolyte composition in accordance with Example 1 of Awano was prepared using Awano's silicon Compound No. 11, which corresponds to Compound No. 10 disclosed in the present application. In this nonaqueous electrolyte composition, a concentration of  $\text{LiPF}_6$ , an electrolyte salt, was 1.5 mol/l, which is the same concentration as in Example 2 described in page 28 lines 10 to 11 of the present specification. Using the thus obtained nonaqueous electrolyte composition, a lithium secondary battery was prepared in the same

manner as described in the present specification at page 22, line 23 through page 23, line 16, and page 23, line 25 through page 24, line 9. The thus obtained battery was evaluated for its initial output, output after 500 cycles, and discharge capacity retention after 500 cycles, each at 20°C and at -30°C, by conducting the tests described in the present specification at page 24, line 17 through page 25, line 22. The results obtained are shown in Table 1 below.

For Comparative Example 3-2, a second nonaqueous electrolyte composition ( $\text{LiPF}_6$  concentration: 1.5 mol/l) this time in accordance with Example 2 of Awano was prepared using Awano's silicon Compound No. 11. Using the thus obtained nonaqueous electrolyte composition, a lithium secondary battery was prepared and evaluated in the same manner described above for Comparative Example 3-1. The results obtained are shown in Table 1 below.

For Comparative Example 3-3, a nonaqueous electrolyte composition ( $\text{LiPF}_6$  concentration: 1.5 mol/l) in accordance with Example 1 of Awano was prepared, this time using Awano's silicon Compound No. 12, which corresponds to Compound No. 13 disclosed in the present application. Using the thus obtained nonaqueous electrolyte composition, a lithium secondary battery was prepared and evaluated in the same manner described above for Comparative Example 3-1. The results obtained are shown in Table 2 below.

For Comparative Example 3-4, a second nonaqueous electrolyte composition ( $\text{LiPF}_6$  concentration: 1.5 mol/l) in

accordance with Example 2 of Awano was prepared using Awano's silicon Compound No. 12. Using the thus obtained nonaqueous electrolyte composition, a lithium secondary battery was prepared and evaluated in the same manner described above for Comparative Example 3-1. The results obtained are shown in Table 2 below.

Examples in accordance with the present invention

Examples 2-3, 2-7, 2-11 and 2-15, as shown in Tables 3 and 4 of the present specification, correspond to results of the above-described tests when performed on a lithium secondary battery prepared using a nonaqueous electrolyte composition comprising silicon Compound No. 10, in accordance with the present invention. The results for Examples 2-3, 2-7, 2-11 and 2-15 as shown in the present specification are provided in Table 1 below.

Examples 2-4, 2-8, 2-12 and 2-16, as shown in Tables 3 and 4 of the present specification, correspond to results of the above-described tests when performed on a lithium secondary battery prepared using a nonaqueous electrolyte composition comprising silicon Compound No. 13, in accordance with the present invention. The results for Examples 2-4, 2-8, 2-12 and 2-16 as shown in the present specification are provided in Table 2 below.

Initial outputs were expressed relatively taking the result of Comparative Example 2-1, as shown in Table 4 of the present specification and reproduced in Tables 1 and 2 below, as 100.

Table 1

			Example				Compara. Example		
			2-3	2-7	2-11	2-15	3-1	3-2	2-1
System of Nonaqueous Electrolyte	Mixed Organic Solvent Formulation (vol%)	EC	30	25	25	25	30	10	30
		EMC	40	40	40	40	40	40	20
		DMC	20	30	25	30	30	30	40
		DEC	10	5	10	-	-	-	10
		PC	-	-	-	5	-	-	-
		BC	-	-	-	-	-	20	-
	Silicon Compound (mol/L)	Compound No.10	0.4	0.4	0.4	0.4	0.4	0.4	-
Results of Measurement	20°C	Initial Output	100.0	100.5	99.8	100.7	99.3	98.2	100.0
		Output after 500 Cycles	94.8	94.8	93.0	93.1	79.8	63.5	76.0
		Discharge Capacity Retention after 500 Cycles (%)	84.3	84.7	83.8	83.9	78.4	64.1	72.9
	-30 °C	Initial Output	135.2	135.0	135.8	134.6	128.5	133.9	100.0
		Output after 500 Cycles	124.1	124.0	125.4	125.6	92.3	79.5	85.0
		Discharge Capacity Retention after 500 Cycles (%)	97.0	96.7	96.7	96.9	89.7	85.4	87.7

Table 2

			Example				Compara. Example		
			2-4	2-8	2-12	2-16	3-3	3-4	2-1
System of Nonaqueous Electrolyte	Mixed Organic Solvent Formulation (vol%)	EC	30	25	25	25	30	10	30
		EMC	40	40	40	40	40	40	20
		DMC	20	30	25	30	30	30	40
		DEC	10	5	10	-	-	-	10
		PC	-	-	-	5	-	-	-
		BC	-	-	-	-	-	20	-
	Silicon Compound (mol/L)	Compound No.13	0.4	0.4	0.4	0.4	0.4	0.4	-
Results of Measurement	20 °C	Initial Output	99.9	100.2	99.6	100.5	99.6	98.4	100.0
		Output after 500 Cycles	94.2	94.3	93.7	93.0	79.9	61.2	76.0
		Discharge Capacity Retention after 500 Cycles (%)	84.9	85.0	84.0	84.7	78.7	62.7	72.9
	-30 °C	Initial Output	134.9	134.7	134.5	133.3	124.4	134.1	100.0
		Output after 500 Cycles	123.0	123.2	123.9	124.8	96.7	88.1	85.0
		Discharge Capacity Retention after 500 Cycles (%)	96.9	96.0	97.0	95.9	88.9	87.3	87.7

As is evident from the foregoing results, the compositions in accordance with the present invention yield lithium secondary batteries having cycle characteristics which are unexpectedly superior to those obtained using the compositions disclosed by Awano. These unexpected superior cycle characteristics are further pronounced at low temperature.

For example, using silicon Compound No. 10 of the present invention (which is Awano's silicon Compound No. 11) at 20°C, the present invention yielded relative outputs after 500 cycles ranging from 93.0 to 94.8, compared with 63.5 and 79.8 obtained with Awano's compositions. At -30°C, these superior characteristics are even more pronounced, ranging from 124.0 to 125.6 using compositions of the present invention compared with 79.5 and 92.3 obtained with Awano's compositions. (See Table 1 above).

Discharge capacity retention characteristics also are unexpectedly superior for the lithium secondary batteries prepared using the compositions of the present invention with silicon Compound No. 10. Specifically, the present invention yielded a superior discharge capacity after 500 cycles ranging from 83.8 to 84.7% at 20°C compared with 64.1 and 78.4% obtained with Awano's compositions, and ranging from 96.7 to 97.0% at -30°C compared with 85.4 and 89.7% obtained with Awano's compositions.



These unexpected superior results are analogously obtained using silicon Compound No. 13 (Awano's silicon Compound No. 12) as is reflected in Table 2 above. Specifically, at 20°C the present invention yielded a relative output after 500 cycles ranging from 93.0 to 94.3 compared with 61.2 and 79.9 obtained with Awano's compositions, and ranging from 123.0 to 124.8 at -30°C compared with 88.1 and 96.7 obtained with Awano's compositions. Likewise, the present invention yielded a superior discharge capacity after 500 cycles ranging from 84.0 to 85.0% at 20°C compared with 62.7 and 78.7% obtained with Awano's compositions, and ranging from 95.9 to 97.0% at -30°C compared with 87.3 and 88.9% obtained with Awano's compositions.

As is reflected in Tables 1 and 2 above, the cycle characteristics obtained using Awano's compositions at both testing temperatures provided only marginal benefit, if any, over the results obtained in Comparative Example 2-1 in which the silicon compound was omitted. In comparison, the compositions in accordance with the present invention consistently provided substantial improvements in the cycle characteristics. This further underscores the result that the superior cycle characteristics obtained using the compositions of the present invention would have been unexpected.

In addition, it is noteworthy that Awano describes silicon Compounds which are represented by general formula (1) of the present invention (see Comparative compounds A and B in U.S.

2004/0007688) and teaches that these compounds do not impart improvements in cyclic and low-temperature characteristics. In contrast, in the compounds according to the present invention, silicon compounds represented by general formula 1 provide superior cycle characteristics that are comparable with the superior results obtained using silicon compounds represented by general formula 2. (See Examples 2-1, 2-2, 2-5, 2-6, 2-9, 2-10, 2-13 and 2-14 in Tables 3 and 4 of the present specification). Awano's contrary results using silicon compounds of general formula 1 even further underscores that the compositions of the present invention provide unexpected superior results and would have been non-obvious over Awano and the other references cited in the Official action.

I further declare that the foregoing statements were made with the knowledge that willful false statements and the like are punishable by fine or imprisonment, or both, under § 1001 of Title 18 of the United States Code and may jeopardize the validity of the application or any patent issuing thereon; and further that all statements made herein of my own knowledge are true and that all statements made herein on information and belief are believed to be true.

Date: November 27, 2009

Takayuki Taki  
(Takayuki TAKI)